Notes on Mercury Emissions and Atmospheric Deposition Within the Lake Superior Basin

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A model-based analysis ("ARL-2005") was carried out to estimate the amount and source-attribution for atmospheric mercury to the Great Lakes with FY2010 funding from the Great Lakes Restoration Initiative (GLRI) (Cohen et al., 2011, 2012). In this short memo, the modeling results have been examined to address the question of emissions and deposition in the Lake Superior Basin (LSB).

1. 2005 Atmospheric Mercury Emissions from Sources in the Lake Superior Basin

The ARL-2005 modeling analysis was carried out for 2005, as this was the year of the latest available U.S. mercury emissions inventory from the National Emissions Inventory (NEI-2005) at the time the analysis was carried out. Large point sources in the U.S. and Canadian inventories used in this analysis are shown in the LSB and surrounding regions in Figure 1 below.

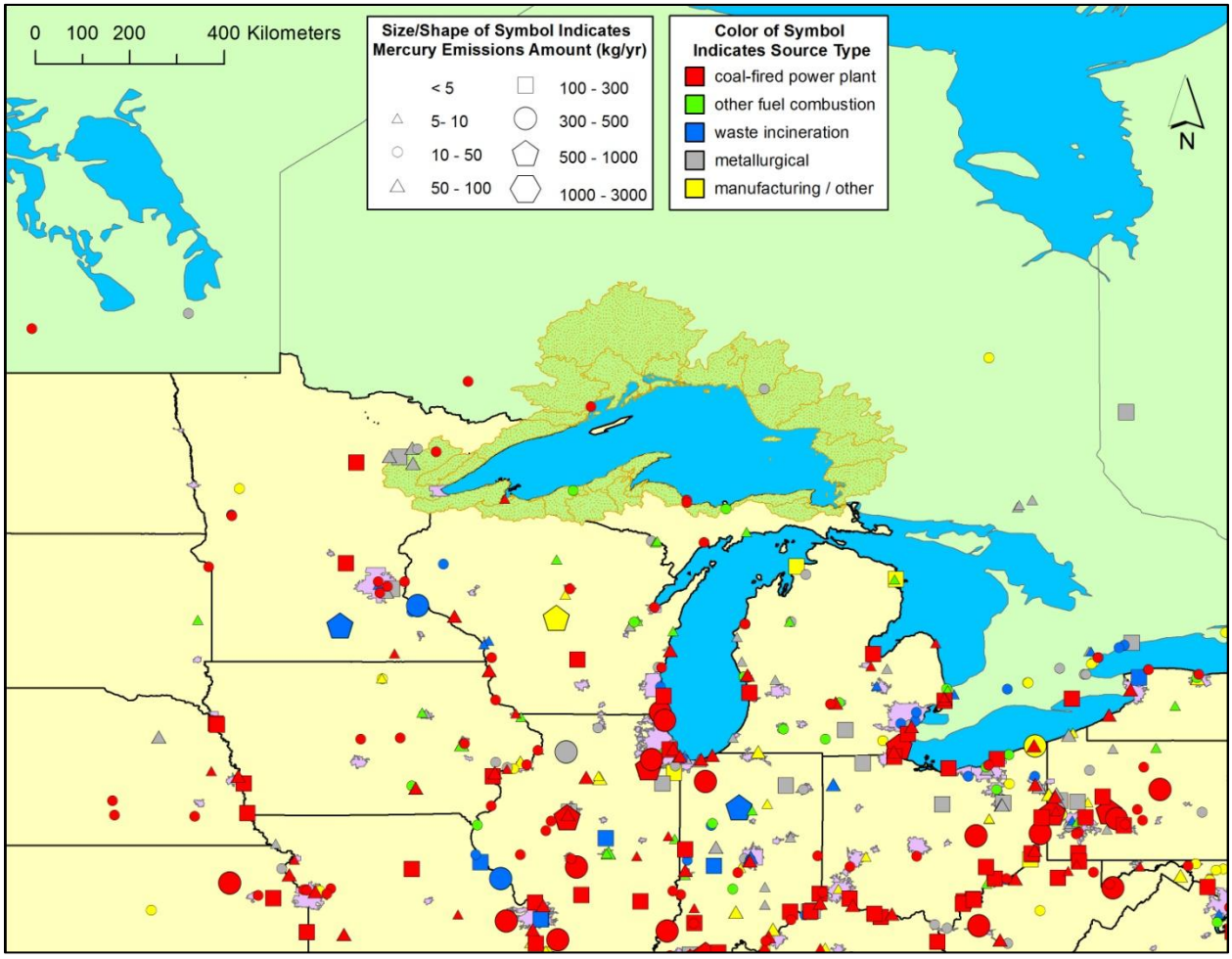

Figure 1. Mercury emissions in the LSB and surrounding regions used in the ARL-2005 modeling analysis

The emissions inventories used in this 2005 analysis were examined to determine which sources were within the Lake Superior Basin, considered here to be the lake and its watershed. The total direct anthropogenic emissions for 2005 – as opposed to natural or mercury re-emitted after being deposited -- within the Lake Superior Basin in the ARL-2005 and the Lake Superior Workgroup estimates were 525 kg/yr and 634 kg/yr, respectively, and are summarized in Figure 2 below.

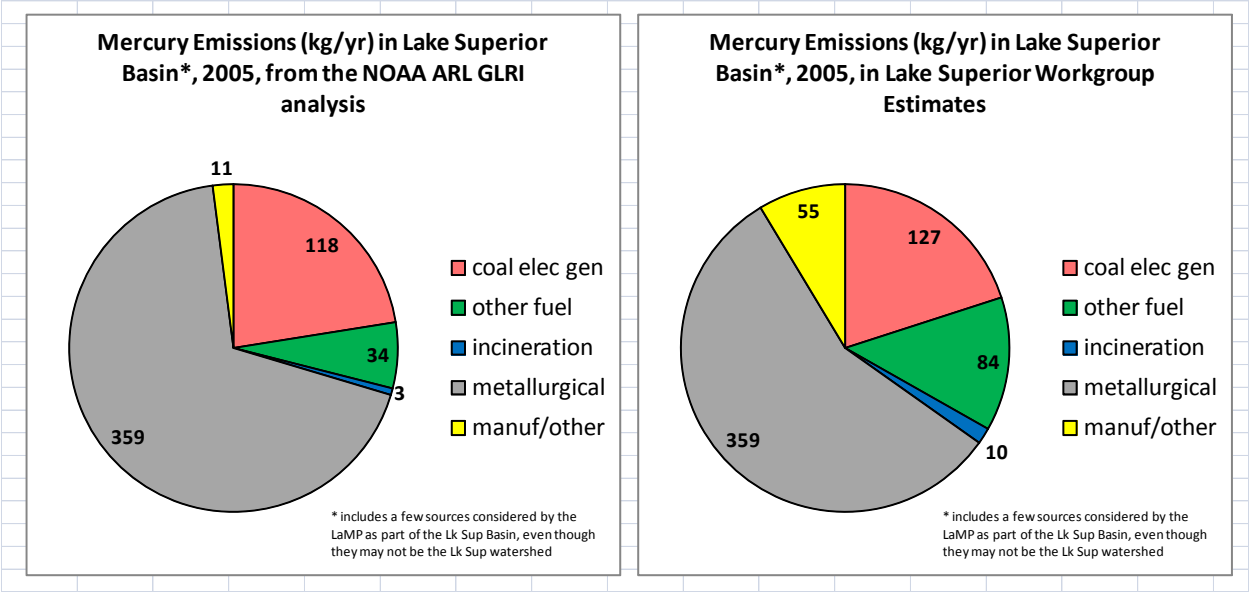

Figure 2. Mercury emissions from LSB sources in the ARL-2005 analysis and estimated from data provided by the Lake Superior Workgroup

Even though there is a difference between the two datasets in LSB mercury emissions estimates, metallurgical facilities appear to be the largest source category of mercury emissions. The two datasets for 2005 point source LSB emissions are reasonably consistent, as they both relied primarily on the USEPA's 2005 National Emissions Inventory (NEI 2005) for U.S. point source emissions and the Canadian 2005 National Pollutant Release Inventory (NPRI 2005) for Canadian point source emissions. As a result, for most point sources the emissions in the two datasets were identical. However, for a few point sources, the estimates were somewhat different. It is beyond the scope of this memo to determine the reasons for the differences, but the primary differences are summarized in Table 1 and Table 2 below. It is seen in Table 1 that for all of the point sources with the largest differences, the Lake Superior Workgroup used a different source of data than the 2005 NEI used by the ARL-2005 analysis. For area sources – summarized in Table 2 – the comparison between the two datasets is difficult to characterize, as very different methodologies were used. For example, the source categories in the Canadian area source inventory used in the ARL-2005 analysis – based on 2000 data provided by Environment Canada – are almost completely different than the categories in the Lake Superior Workgroup estimates. For U.S. area sources, the ARL-2005 analysis used data from 2002 NEI. In contrast, the Lake Superior Workgroup estimates used different estimation methodologies and included additional area source categories relative to the 2002 NEI.

Table 1. Largest differences in LSB point source mercury emissions between ARL-2005 analysis and Lake Superior Workgroup estimates for 2005

Facility Name	ARL-2005 analysis		Lake Superior Workgroup estimates		Difference
	2005 Mercury Emissions (kg/yr)	Data source	2005 Mercury Emissions (kg/yr)	Data source	2005 Mercury Emissions (kg/yr)
Taconite Harbor Energy Center	0.18	NEI 2005	25.81	"See 'MPCA Hg Est – Facilities' tab"	**+25.63**
Stone Container Corporation	0.75	NEI 2005	10.80	2005 TRI	**+10.05**
Xcel Energy Bay Front Generating Station	6.37	NEI 2005	13.91	Baudhuin, Neal Neal.Baudhuin@dnr.state.wi.us	**+7.54**
Minnesota Power Inc. - M.L. Hibbard	-	Apparently not included in NEI 2005	2.72	"See 'MPCA Hg Est – Facilities' tab"	**+2.72**
Graymont (WI) Inc.	-	Apparently not included in NEI 2005	2.59	2008 TRI	**+2.59**
St. Louis County Regional SW	-	Apparently not included in NEI 2005	1.71	2002 NEI	**+1.71**
Sappi Cloquet LLC	0.63	NEI 2005	2.25	12/21/10 e-mail from Justin Finke, Sappi	**+1.63**
MARQUETTE BOARD OF LIGHT and POWER	10.80	NEI 2005	8.16	2003 TRI	**-2.64**
Minnesota Power Inc - Laskin Energy Ctr	14.10	NEI 2005	9.53	"See 'MPCA Hg Est – Facilities' tab"	**-4.57**

Table 2. Overall differences in LSB area source mercury emissions between ARL-2005 analysis and Lake Superior Workgroup estimates for 2005

Source Category	ARL-2005 analysis		Lake Superior Workgroup estimates		Difference
	2005 Mercury Emissions (kg/yr)	Data source	2005 Mercury Emissions (kg/yr)	Data source	2005 Mercury Emissions (kg/yr)
U.S Area Sources in LSB	6.64	NEI 2002	44.3	Numerous worksheets within the U.S. LSB emissions spreadsheet	**+37.66**
Canadian Area Sources in LSB	10.15	Environment Canada, data for 2000, personal communication	20.9	Numerous worksheets within the Canadian LSB emissions spreadsheet	**+10.75**

2. 2005 Atmospheric Mercury Deposition from Sources in the Lake Superior Basin

As described in Cohen et al (2011, 2012), the NOAA HYSPLIT-Hg model was used to estimate the atmospheric mercury deposition contribution of each source in the inventories used to each of the Great Lakes and their watersheds. The results for Lake Superior (and its watershed) were extracted from the analysis and will be summarized briefly here. Summing up the individual deposition contribution results for each of the sources in the LSB used in the ARL-2005 analysis, LSB sources were estimated to contribute a total of 9 kg/yr of mercury via atmospheric deposition to Lake Superior directly, 20 kg/yr to the watershed, and 29 kg/yr to the combined watershed and lake. A breakdown of the relative contribution of different source types to these deposition contributions is shown in Figure 3. It can be seen from this figure that coal-fired power plants appear to be the source category making the largest atmospheric deposition contribution, to the lake, watershed and basin.

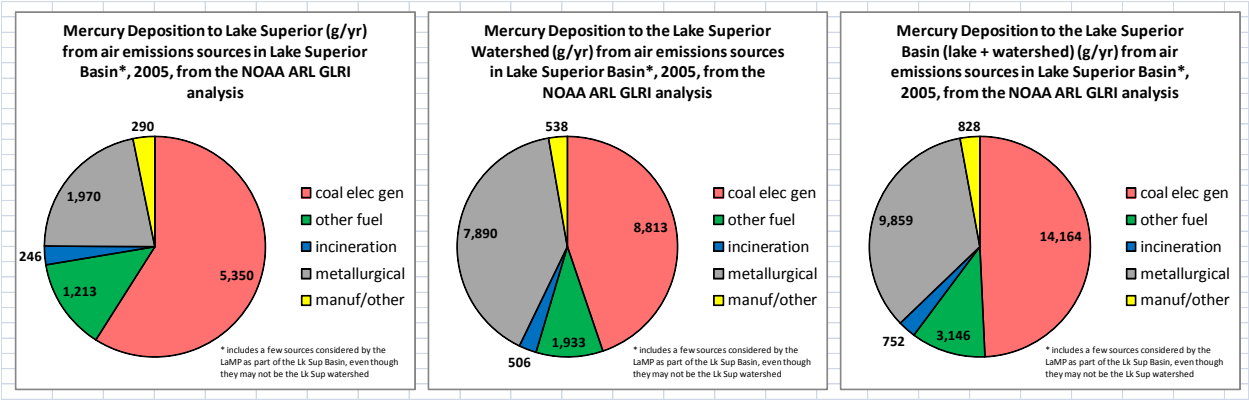

Figure 3. Atmospheric mercury deposition from LSB mercury emissions sources to Lake Superior itself, the Lake Superior watershed, and the Lake Superior Basin (lake + watershed), based on the ARL-2005 analysis

This result can be contrasted with the emissions data presented in the previous section which showed that metallurgical operations were the largest *emissions* source in the Lake Superior Basin. What are the reasons for this difference? Examination of the map in Figure 1 suggests that the locations of the different source types within the LSB may play a role, as the coal-fired power plants within the LSB tend to be closer to the lake than the metallurgical facilities. Another reason may be the different speciation of emissions from the two sectors. By "speciation" we mean the relative proportion of different forms of mercury: elemental, reactive gaseous, and particulate. The estimated speciation of emissions for the different source sectors in the Lake Superior Basin is shown in Figure 4. It can be seen that metallurgical emissions are largely in the elemental mercury form [Hg(0)]. In contrast, coal-fired power plant emissions include a more significant fraction of reactive gaseous mercury (RGM). Because it wet and dry deposits much faster than Hg(0), RGM is believed to have a much greater "local" and "regional" deposition impacts. So, even though coal-fired power plants were estimated to emit less than metallurgical facilities, a higher fraction of the power plant emissions were in a form that deposited more readily within the LSB.

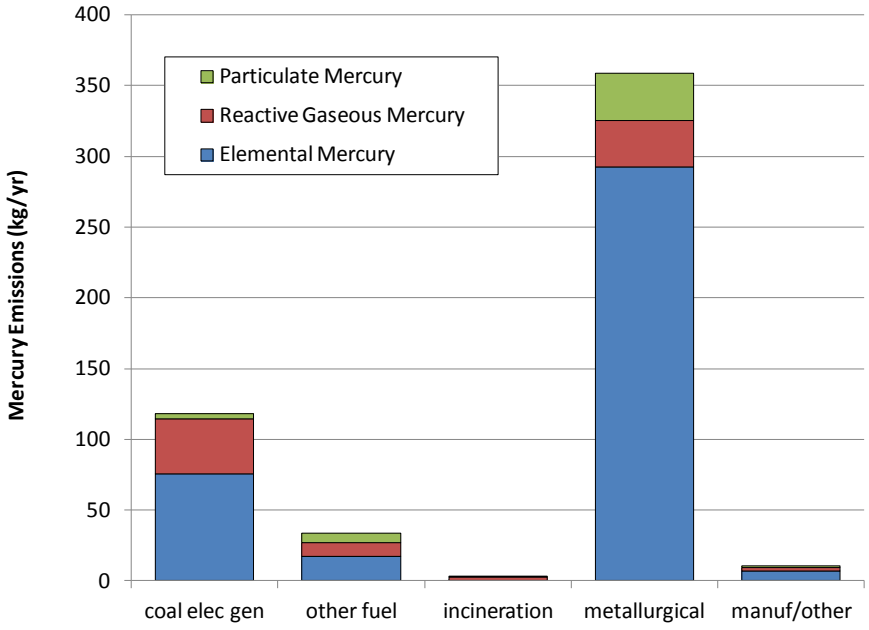

Figure 4. Estimated speciation for LSB mercury emissions sources, based on ARL-2005 analysis

3. 2005 Atmospheric Mercury Emissions and Deposition from Sources Inside and Outside the Lake Superior Basin

Of course, it is realized that sources outside the Lake Superior Basin can contribute atmospheric mercury deposition to the Lake Superior ecosystem. Based on the same analysis as described above, the overall emissions and deposition from different source types – for sources both within and outside of the LSB – are summarized in Figure 5, Figure 6, Figure 7, and Figure 8 below. It can be seen that the total emissions and deposition arising from LSB sources are a very small fraction of the total. The total emissions from all sources within the LSB are on the order of 0.015% of the total global emissions, and the deposition to the LSB from these sources are on the order of 1.3% of the total contribution from all global sources. The deposition from these LSB sources is a much higher fraction of global deposition than the emissions (1.3% vs. 0.015%, or more than a factor of 100), but the overall impact of the these LSB sources on the LSB is very small.

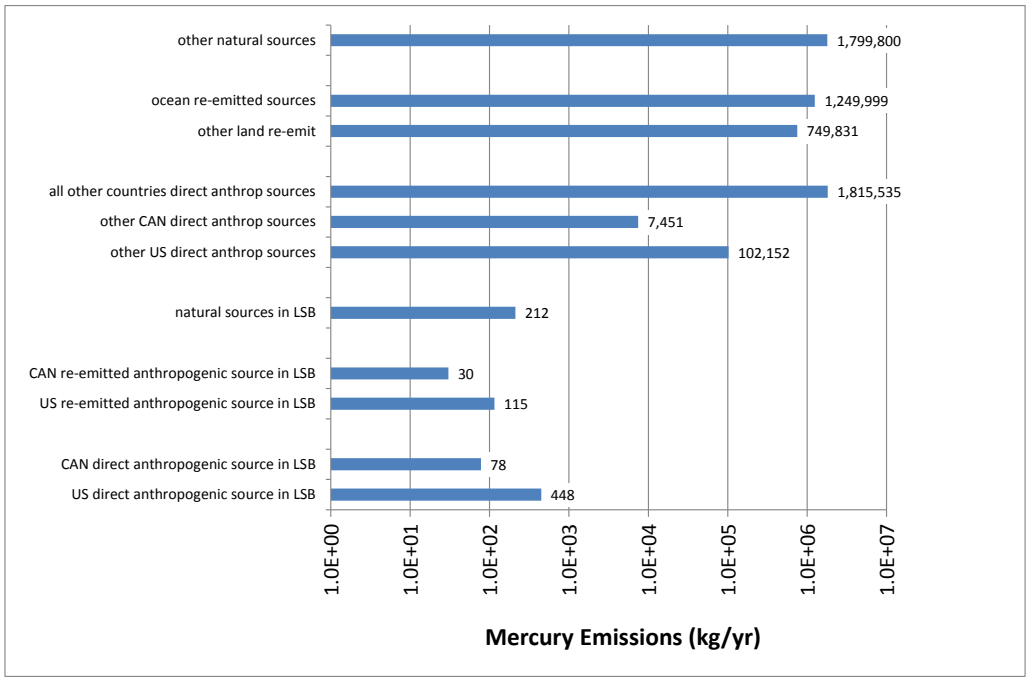

Figure 5. Mercury emissions from sources within the LSB and outside the LSB, based on the ARL-2005 analysis

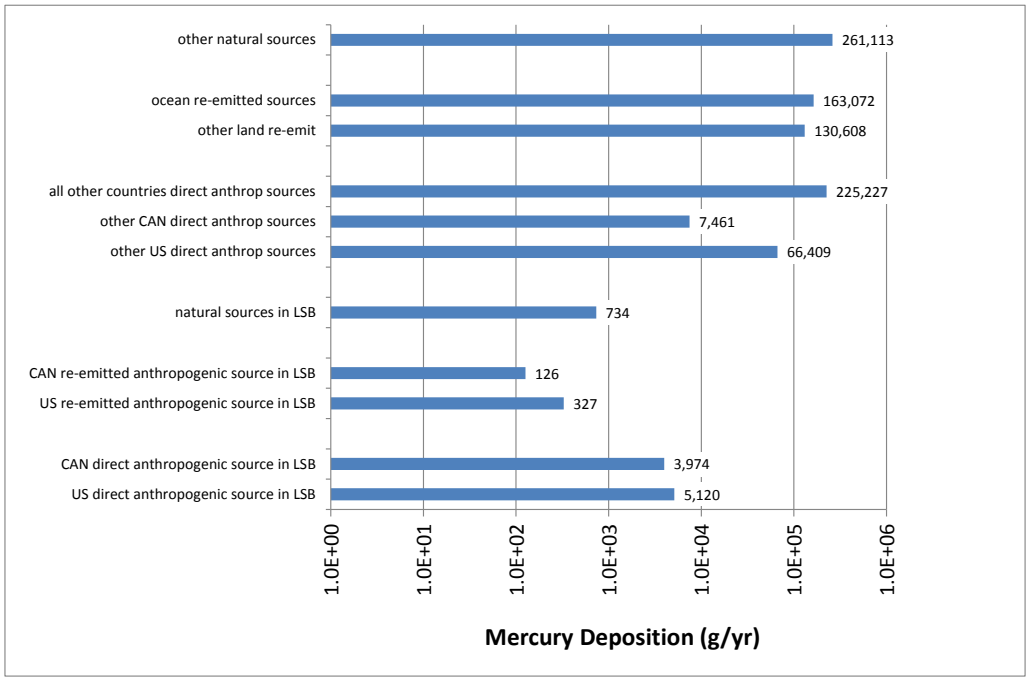

Figure 6. Atmospheric mercury deposition to Lake Superior from sources within & outside the LSB, based on the ARL-2005 analysis

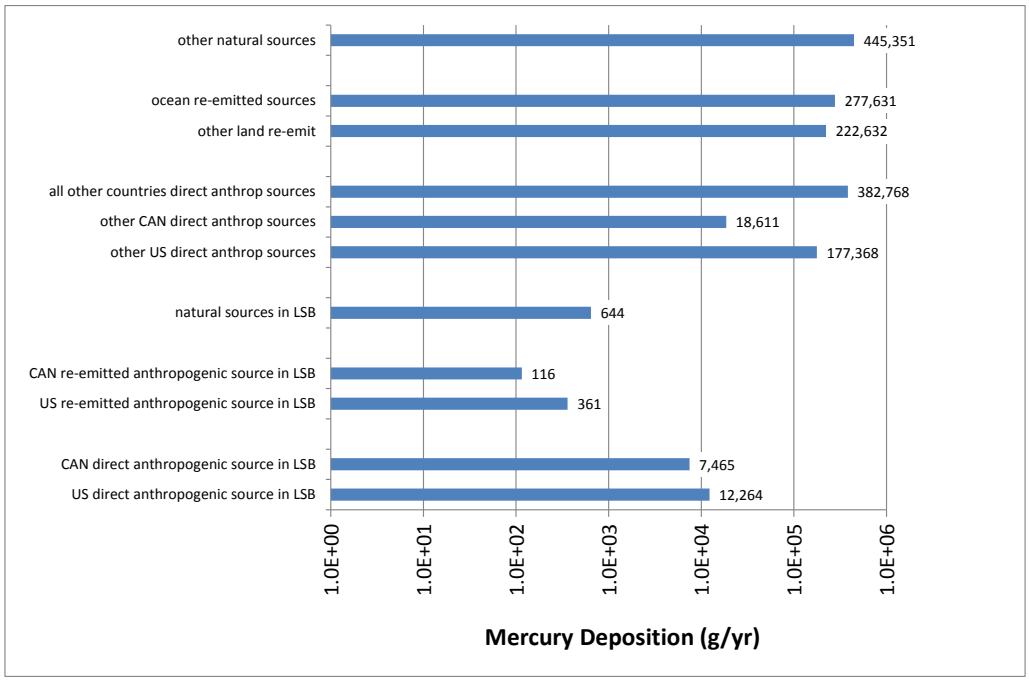

Figure 7. Atmospheric mercury deposition to the Lake Superior watershed from sources within & outside the LSB, based on the ARL-2005 analysis

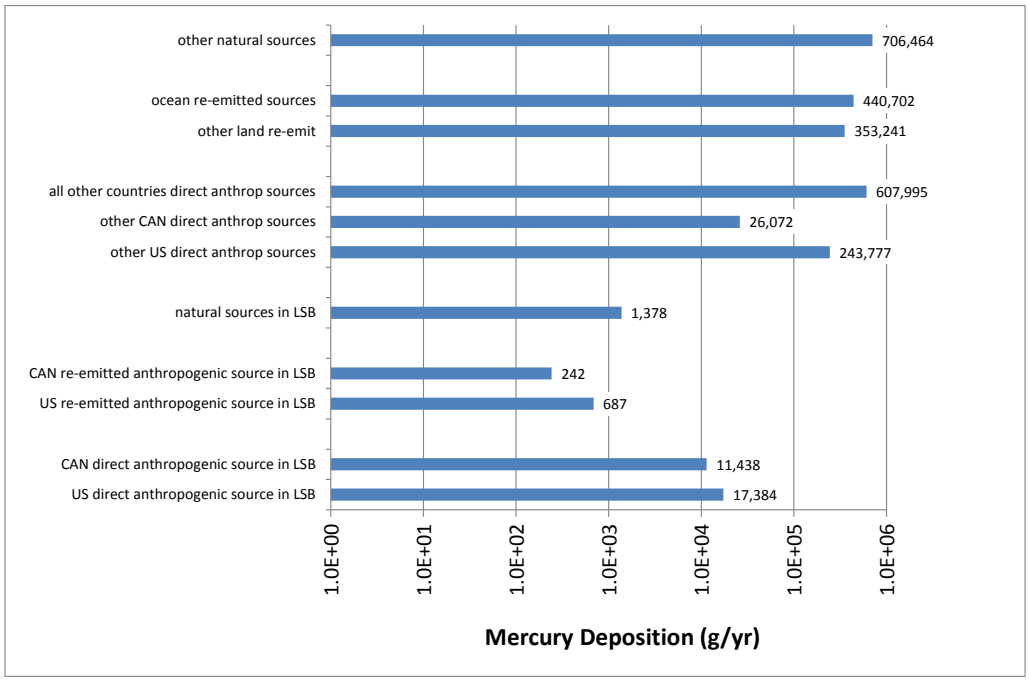

Figure 8. Atmospheric mercury deposition to the Lake Superior Basin (lake + watershed) from sources within & outside the LSB, based on the ARL-2005 analysis

4. 2010 vs. 2005 Atmospheric Mercury Emissions in the Lake Superior Basin

As mentioned above, the ARL-2005 modeling analysis was carried out for 2005, as that was the most recent year for which the U.S. EPA's National Emissions Inventory (NEI) was available for mercury emissions. All of the results above are related to this 2005 analysis. Of course, emissions from sources have changed since 2005. The Lake Superior Workgroup has generated estimates of 2010 emissions from LSB sources that can be compared with estimates of 2005 emissions. In Table 3, the sources with the largest differences – positive or negative – between the 2010 and 2005 estimated emissions are shown. It can be seen from this table that most of the largest changes represent a decrease in emissions between 2005 and 2010. In these sources with the largest changes, emissions from LSB coal-fired power plants were estimated to have decreased by about ~55 kg/yr – representing nearly half of the total emissions from this source category in the LSB -- and emissions from metallurgical facilities (including mining) were estimated to have decreased by about ~29 kg/yr – representing only about ~10% of the emissions from this source category in the LSB.

Significant changes in emissions are usually accompanied by changes in emission speciation. This is because changes in pollution control equipment and/or production processes tend to change the relative proportions of different forms of mercury in the air emissions from sources. At the same time, changes in emissions speciation can have a dramatic impact on the geographical distribution of atmospheric deposition in the region around the source. It is beyond the scope of this memo to attempt to estimate the 2010 speciation and "re-estimate" the associated deposition for each source with major emissions changes. However, if changes in emissions from coal-fired power plants resulted in significant changes in RGM emissions – the type of mercury that deposits more readily – then the deposition impacts from this category might be decreased significantly. If this were the case, then the relative contribution from metallurgical and coal-fired power plants would change. The finding above – that LSB coal-fired power plants were estimated to contribute more to the LSB than metallurgical operations (for the ARL-2005 analysis) – would thus be changed for the 2010 situation. Given the relative magnitude of the two contributions in 2005 and the relative magnitude of the emissions changes, it is likely that the contribution of both source categories to the LSB would be roughly comparable. A more detailed analysis of this more current situation could certainly be carried out.

Table 3. Largest Differences between 2010 and 2005 emissions estimates for LSB sources

Facility Name	Source type*	Lake Superior Workgroup estimates		Lake Superior Workgroup estimates		Difference
		2005 Mercury Emissions (kg/yr)	Data source	2010 Mercury Emissions (kg/yr)	Data source	2010 – 2005 Mercury Emissions (kg/yr)
Northshore Mining Co - Silver Bay	**4**	4.67	"See 'MPCA Hg Est – Facilities' tab"	21.22	"See 'MPCA Hg Est – Facilities' tab"	**+16.5**
Minnesota Power Inc. - M.L. Hibbard	**1**	2.72	"See 'MPCA Hg Est – Facilities' tab"	7.17	"See 'MPCA Hg Est – Facilities' tab"	**+4.5**
TACONITE HARBOR ENERGY CENTER	**1-2**	25.81	"See 'MPCA Hg Est – Facilities' tab"	28.71	"See 'MPCA Hg Est – Facilities' tab"	**+2.9**
WISCONSIN ELECTRIC POWER COMPANY	**1**	55.47	2005 TRI	13.70	2009 TRI	**-41.8**
US Steel - Keewatin Taconite	**4**	66.63	"See 'MPCA Hg Est – Facilities' tab"	47.99	"See 'MPCA Hg Est – Facilities' tab"	**-18.6**
Williams Mine	**4**	21.31		3.5	2009 data used	**-17.8**
WHITE PINE ELECTRIC POWER LLC	**1**	14.15	2005 TRI	0.0036	2009 TRI	**-14.1**
STONE CONTAINER CORP	**5**	10.80	2005 TRI	-	No data given?	**-10.8**
Area source: Residential Oil Combustion, Minnesota	**2**	9.49	NEI derived - See "Residential Fuel" tab	2.41	2008 NEI - See NEI calcs	**-7.1**
Thunder Bay Generating Station	**1**	37.2		31	2009 data used	**-6.2**
NEWMONT CANADA LIMITED - Golden Giant Mine	**4**	4.673		-		**-4.7**
BOWATER - Thunder Bay Operations	**5**	4		0		**-4.0**
US Steel Corp - Minntac	**4**	87.95	"See 'MPCA Hg Est – Facilities' tab"	84.05	"See 'MPCA Hg Est – Facilities' tab" + NEI 2005	**-3.9**

* Source type codes: 1 = coal-fired power plants; 2 = other fuel combustion; 3 = waste incineration; 4 = metallurgical; 5 = manufacturing and other

5. Notes Regarding Methodology

As described in Cohen et al. (2011, 2012), the modeling analysis is based on a set of unit-emissions simulations from "hypothetical standard source locations" (HSSL's). From the results of these simulations, spatial and chemical interpolation procedures are used to estimate the impact of each actual source in the emissions inventories used in the analysis. Due to computational resource restraints, the number of HSSL's must be limited. The choice of the location and number of HSSL's is determined by a number of factors. Ultimately, the goal is to use enough HSSL's to ensure that the interpolation procedures can produce a reasonably accurate estimate of the amount and source attribution of mercury deposition to receptors of interest. So, HSSL's are not generally located in regions with few sources. From the map shown above in Figure 1 it is seen that there are few large sources in the Lake Superior Basin. Based on this, there were very few HSSL's chosen in this region. As noted above, only about 1% of the deposition to the LSB was estimated to arise from these sources, and so, significant computational resources were not expended on sources in the LSB. A map of HSSL's used in the ARL-2005 modeling analysis is shown in Figure 9 below. It can be seen that there indeed only a few HSSL's in the LSB. Comparing the locations of the largest LSB emissions sources (Figure 1) with the locations of the LSB-relevant HSSL's shows that these HSSL's will likely provide a reasonably accurate basis for interpolation. However, a more accurate assessment of the contribution of LSB sources could be made if more HSSL's were added. Indeed, simulations could be made for every source in the region, if resources were available. While the accuracy would be increased with more HSSL's, it is not believed that the results will change qualitatively, i.e., the overall amount of and source-attribution for LSB mercury deposition will not change dramatically.

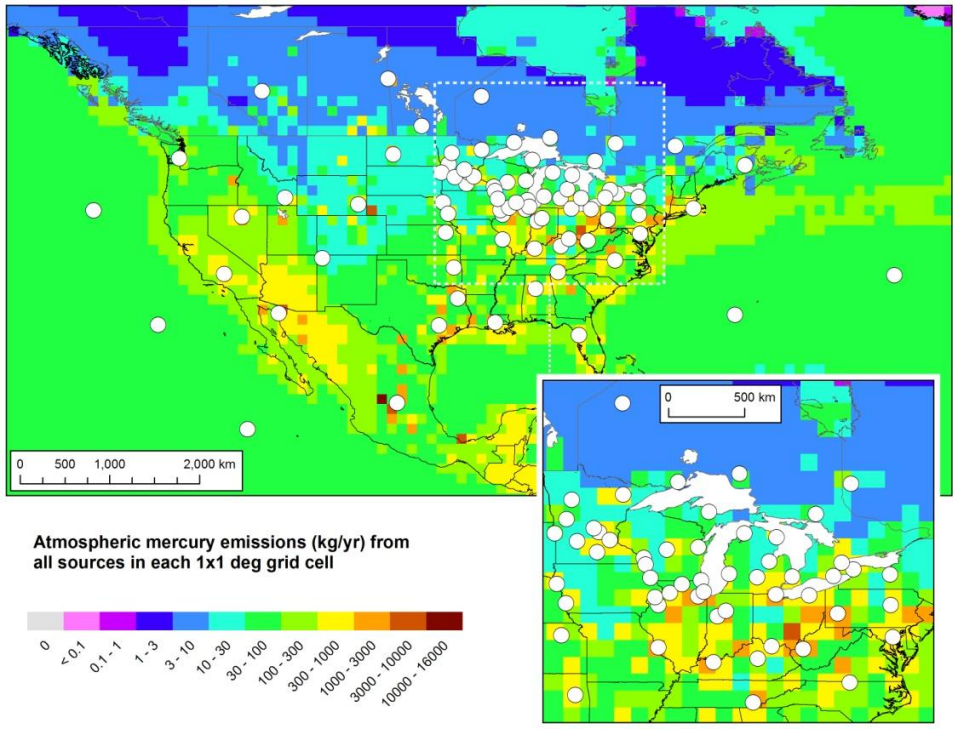

Figure 9. Hypothetical Standard Source Locations (HSSL's) in North America used in the ARL-2005 modeling analysis

References

Cohen, M., Draxler, R., Artz, R. (2011). Modeling Atmospheric Mercury Deposition to the Great Lakes. Final Report for work conducted with FY2010 funding from the Great Lakes Restoration Initiative. December 16, 2011. NOAA Air Resources Laboratory, Silver Spring, MD.

Report (160 pages):
http://www.arl.noaa.gov/documents/reports/GLRI_FY2010_Atmospheric_Mercury_Final_Report_2011_Dec_16.pdf
Figures/Tables in PowerPoint:
http://www.arl.noaa.gov/documents/reports/Figures_Tables_GLRI_NOAA_Atmos_Mercury_Report_Dec_16_2011.pptx
One-page summary (with figures):
http://www.arl.noaa.gov/documents/reports/GLRI_Atmos_Mercury_Summary.pdf

Cohen, M., Draxler, R., Artz, R. (2012). Modeling the Atmospheric Deposition of Mercury to the Great Lakes (work funded through the Great Lakes Restoration Initiative). NOAA Air Resources Laboratory, Silver Spring, MD, USA. Presentation to the IJC's International Air Quality Advisory Board. April 25, 2012, Washington D.C. http://www.arl.noaa.gov/documents/reports/Cohen_IAQAB_Apr_25_2012.pptx